

## **REMARKS**

The Examiner's comments together with the cited references have been carefully studied. Favorable reconsideration in view of the foregoing amendments and following remarks is respectfully requested. Applicants thank the Examiner for the indication of allowable subject matter.

New claims 25 and 26 have been added to provide coverage directed to alternative embodiments of the invention.

Applicants herewith file a terminal disclaimer in compliance with 37 CFR 1.321(c) to overcome a provisional double patenting rejection based on a nonstatutory double patenting ground.

Claims 19-21 stand rejected under 35 U.S.C. 112, second paragraph. The Examiner states that the use of the term, "chelating agent" in the claims is erroneous and misleading. The Examiner states that the term is misleading as the meaning has not been identified and it is not used in its ordinary meaning. This rejection is respectfully traversed. Claim 18 has been amended to incorporate language from page 8 lines 18-21 of the specification and to omit specific reference to the term "chelating agent." The rejected claims now refer to the modifying agent as described in the specification. While not necessary in view of this amendment to claim 18, it is further pointed out that the use of the term "chelating agent" such as utilized in this application does have basis in the literature such as GB Deacon and R. J. Philip's, Coordination Chemistry Reviews, 33 (1980), pg 227-250, although it is likely "complexing," agent would be the more commonly used term. In any event, in view of the modification of claim 18, it is respectfully urged that the claims are definite, and reconsideration and withdrawal of this rejection is respectfully requested.

Claims 1-8 and 10-24 stand rejected under 35 U.S.C. 103(a) as being unpatentable over Poncelet (US 6,468,492) in light of Pinnavaia (US 10/272,751). The rejection is traversed. A key reason why the present invention is believed patentable over the art is the fact that it is conducted at an ambient temperature of 15°C to 35°C. It was to preclude higher temperature heating that the term "directly" was inserted at the end of then step (c) in the prior amendment and is maintained in step b) of this amendment. Before dealing with the art rejection, the enclosed Declaration Under Rule 132 will be discussed.

The enclosed Declaration Under Rule 132 by co-inventor Desrousseaux confirms that the product of the claimed method of the invention is materially altered if the temperature is elevated above ambient. Clearly, the process of the '646 patent calls for an intentional heating and produces a crystalline product. The present invention calls for ambient temperature and results in an amorphous product. Although said to be at a temperature less than 100°C, there is clearly an affirmative heating step in '646. An affirmative heating step clearly excludes ambient temperature ( which by definition means no affirmative heating step). The example of the '646 patent is conducted at 96-98°C, which would not be regarded as ambient by anyone. The subject of the '646 patent is described as a better way of making a filamentous tubular crystallized aluminosilicate. The Declaration at page 2 confirms that the reference spectra (as observed by Transmission Electronic Microscopy, on electronic diffraction patterns and by the presence of sharp bands on the Raman Spectra) are crystalline, tubular filamentary materials. Although the Examiner suggests that the product of the instant invention is not unexpected, it is established by the Raman spectra that the product of the present invention is amorphous, unlike that of the reference. Accordingly, it is demonstrated that the temperature difference provides a significantly different resulting structure and is thus unexpected. Thus, the Examiner's assertion that the '646 patent suggests a similar process are overcome.

Returning to the art rejection:

The Examiner sets forth in his rejection the teachings of the '646 patent in great detail but does not adequately consider the heating requirement and the 96-98 degree temperature of the examples.

Poncelet ('646) teaches a method of preparing an aluminosilicate polymer of the imogolite type having a filamentous tubular crystallized structure. In order to prepare such a specific structure it is required that the method includes a solubilization, ripening, and thermal treatment step.

Thus, the first step is, (as in the present invention), a step of contacting a mixed aluminium-silicon alkoxide substituent or a precursor thereof with an aqueous alkali, keeping certain concentrations within set limits. As shown in the Example, this will normally be followed by a solubilization step, for example, with an acid such as acetic acid, to decrease the pH and solubilize the

precipitate. Then a ripening step at room temperature is required which according to col. 2 line 47 takes at least 5 days and enables the time for the subsequent thermal step to be considerably reduced. The mixture then has to be heated at a temperature "below 100°C" and according to col. 2 lines 63-64 at 96-98°C for 24 hours, which results in the "formation in solution of an aluminosilicate polymer of the imogolite type" (see col. 3 lines 1 and 2). Finally, as with the present invention, residual ions from the polymer are removed.

However, the Examiner has already appreciated that the process of the present invention differs from that of Poncelet ('646) in that, unlike in the latter, it is specifically required that the silicon has both hydrolyzable substituents and a non-hydrolyzable substituent and, with respect to claims 18-22, that the addition of chelating agent occurs after the purification of the product. A fundamental distinction is that, in Poncelet ('646), there is no teaching or suggestion that would lead the skilled man to expect that he should omit the heating step and that the whole reaction could be conducted at ambient temperature and result in a different product.

It should further be pointed out that the material described in Poncelet ('646) is a crystallized aluminosilicate, designated imogolite (col. 1, line 14). This material has an anisotropic, tubular, filamentous structure, made of a bent gibbsite-like sheet (aluminum hydroxide material) forming the exterior part of the filamentous particle and a silica monolayer, covalently linked to the gibbsite-like sheet, forming the interior part of the filamentous particle.

The material described in the present invention exhibits an isotropic structure with, however, a mixture of the silanol and organic moiety on the interior part of the structure, but has a non-filamentous, spherical morphology and is amorphous in character, as shown by electron diffraction (see page 3, line 14).

Indeed the specification discloses that the material obtainable by the method in accordance with the method of Poncelet ('646) is quite different from that of the present invention, as can be seen by a comparison of the Raman spectra. Thus Comparative Example 1 is as described in EP-A-1 112 959 which is the equivalent of Poncelet ('646). It is said at the end of this Example that materials that have the same Raman signature belong to the same family.

The Raman spectrum of this material is represented by Fig. 1 and it is stated on page 9, lines 20-21 that an imogolite fibrous aluminosilicate polymer was obtained. The relevant footprint for comparison is at the low wave number, the peak at the higher wave number end corresponding with the presence of the carboxylate ion from the addition of the acetic acid in step (a). It will be seen that there is a main peak at about 500  $\text{cm}^{-1}$  with a peak close on either side.

In order to make as close a comparison as possible with a product obtained in accordance with the invention, this should be compared with Fig. 9 as this pertains to Example 10 which included the addition of acetic acid, albeit as a chelating agent in step e). Again there are peaks corresponding to the acid at the high wavenumber, but it will be clearly seen that the Raman spectrum is quite different as befitting a very different structural material. Here the main peak is at 1000  $\text{cm}^{-1}$ , but there is no discernible peak at 500  $\text{cm}^{-1}$ , confirming the very different nature of the spherical, amorphous aluminosilicate. Thus the materials produced according to the process of Poncelet ('646) on the one hand and the present invention on the other hand are quite different, that of Poncelet ('646) being in a fibrous, tubular form and that of the present invention being in a spherical, amorphous form. As a different product is produced, it is further teaching that the method steps selected for the instant invention are not obvious.

The Examiner however has stated that, although Poncelet ('646) does not use a silicon compound having a non-hydrolyzable precursor, he considers that use of such a compound is taught however in Pinnavaia (165) and that there is significant motivation to combine the disclosure of Poncelet ('646) with that of Pinnavaia (165).

The anisotropic structure according to the process of Poncelet ('646) is obtained thanks to the  $\text{sp}^3$  character of the silicon atom and to the covalent interactions of the silicon atom with the aluminum atom through three oxo-bridges. The key parameter which allows this kind of isotropic structure to be synthesized is the control of the curvature of the proto-gibbsite sheet by matching the reactivity of both gibbsite and silica precursors: if the synthesis does not allow the control of this crucial step, the final product will be a mixture of gibbsite and silica or organosilica instead of the required aluminosilicate structure.

Pinnavaia (165) teaches the use of silane coupling agents having hydrolyzable groups and one, two, or three non-hydrolyzable groups (see col.11, lines 46-52) without distinguishing between these options. However the reactivity of silica precursors such as silane coupling agent  $R_xSi(OR')_y$  ( $x=0-4$  and  $y=4-0$ , with  $x+y=4$ ) is strongly affected by the nature and the number of R and R' groups and many of these coupling agents could not be used in the present invention, due to the reactivity mismatch between the gibbsite precursor and the silica precursor.

The Poncelet ('646) reference does not disclose (1) the use of a silicon compound having a non-hydrolyzable substituent as a precursor, (2) the adding of alkaline to increased alkali/aluminum molar ratio to 3, (3) directly preceding to eliminate byproducts formed during formation steps from the reaction mixture without a heating step as in Poncelet ('646), and (4) the claim 18-22 use of chelating agents after the purification of the product. As stated above it is respectfully urged that Pinnavaia (165) does not supply teachings that will allow one of ordinary skill in the art to reach the instant invention.

Thus, it has been found that only silane coupling agents bearing one non-hydrolyzable group (R) with the other groups being hydrolyzable groups (R') can be used in the present invention. Other silane coupling agents with more non-hydrolyzable groups e.g.  $R_xSi_x(OR')_y$ , ( $x=2$  or  $3$ ,  $y=2$  or  $1$  and  $x+y=4$ ) would not lead to the silica composition having the structure described in the present invention, because there would not be the three necessary oxo-bridges involved in the mechanism to form the anisotropic structure, coming from the three hydrolyzable groups.

There was no suggestion from Pinnavaia (165) that selection of a coupling agent with only one non-hydrolyzable group would lead to the synthesis of the structure described in the present invention. Nor is there any suggestion that a silane coupling agent without a non-hydrolyzable group, as in the invention of Poncelet ('646), could be used. There was thus no motivation to combine the disclosures thereof.

In fact it will be appreciated that the invention of Pinnavaia (165) relates to a method of preparing a lamellar, mesoporous quasi-crystalline silica composition by the use of a gemini amine surfactant acting as a template or structure directing agent when reacting an orthosilicate. The surfactant can later

be removed, leaving the structural framework, and the resulting composition can react with functional groups to extend the utility of the compositions, for example as metal ion traps for the removal of toxic metals from contaminated water.

The use of a surfactant is thus an essential part of the invention for forming the structure of the composition of Pinnavaia (165). In contrast there is no surfactant used or necessary in preparing either the composition of the present invention or that of Poncelet ('646) and thus there is again no motivation for the skilled man to consider combining the disclosures of Poncelet ('646) and Pinnavaia (165). The skilled person would know that in Pinnavaia (165) the non-hydrolyzable constituent borne by the silane coupling agent would interact with the surfactant and help to grow the expected aluminosilicate structure. In the present invention, however there is no expectation that the non-hydrolyzable group of the silane coupling agent would help to grow the isotropic structure.

The Examiner states that the present invention is distinguished from Poncelet ('646) in that it teaches the introduction of a 'chelating agent' after the purification of the product. According to Poncelet ('646), a 50:50 mixture by volume of HCl and  $\text{CH}_3\text{CO}_2\text{H}$  as chelating agent is used immediately after step (a), its purpose being only to redisperse, i.e. solubilize, the precipitate by lowering the pH which is necessary before the ripening step. One would not expect the desired result because of the low pH value.

In contrast, the chelating agent, if used, is for quite a different purpose in the present invention, being an optional final step, i.e. after eliminating the byproducts. The purpose of the chelating agent here is to allow the modification of the surface of the aluminosilicate polymer by forming a chelate compound, the functional group of the chelating agent allowing an increase in affinity of the aluminosilicate polymer with the medium in which it is used (as explained on page 8, lines 18-21). The chelating agents are used at entirely different steps in the overall process for different purposes and as shown above result in the preparation of materially different products.

The Examiner refers to Pinnavaia (165) as teaching that surfaces may be functionalized by reaction with various chlorides, fluorides, silylating or alkylating agents. In contrast it will be seen that the chelating agents used in the present invention are acids such as carboxylic acids, phosphonic acids, esters, amino acids etc. There is no hint, teaching, or suggestion in Pinnavaia (165) that

such a ‘chelating agent’ was to be used, nor that it could be used in association with the multistep process of the present invention.

**Response to the Examiners Comments on the Applicant’s Argument**

In paragraph 1 bridging pages, 11-12 of the Office Action the Examiner urges that the method used by Poncelet (‘646) has an optional heating step. Poncelet never characterizes the heating as optional. It is not a reasonable interpretation of the English language to say that “heating to a temperature less than 100°C” includes not heating at all. Further, Poncelet (‘646), always involves heat as in the 90°C mixing and aging step. It is respectfully urged that the specification and Examples of the instant application indicate that no heating is carried out. The language that the method proceeds “directly” to the eliminating of byproducts is intended to exclude a heating step. The Examiner also comments that deleting the word “optional” would better differentiate the current claim from prior art. However, it is believed that the present claims adequately distinguish over the art.

In paragraph 2 on page 12 it is gratefully noted that the Examiner indicates that claim 24 would be allowable if incorporated into Claim 1. Nevertheless, it is respectfully urged that the claimed method of the instant application is different from Poncelet (‘646) as admitted by the Examiner, and further that there is no teaching to make a combination with Pinnavaia (165) to modify the teaching of Poncelet (‘646) to overcome the differences as set forth above. Therefore, it is urged that all claims are allowable.

In paragraph 3 bridging pages 12 and 13 of the Office Action, the Examiner urges that while the Pinnavaia (165) reference does not point out an exact number of hydrolyzeable groups on the silane, but sets forth silane precursors having only one non-hydrolyzeable group are well-known and that their use would be obvious. However, while Examiner is correct that one non-hydrolyzeable group site silanes are known, that is not really the issue. The issue is whether it is obvious from the known silanes to pick the right specific silane out of those possibly available for use in modifying the Poncelet (‘646) teachings. It is respectfully urged that there is no suggestion to pick any silane much less the one non-hydrolyzeable silane as claimed in this invention.

The Examiner’s, arguments in paragraphs 4 and 5, page 13 of the Office Action deal with the chelating and functionalizing of the alumino-silicate.

The invention of Pinnavaia (165) is very different from that of either Poncelet ('646) or the present invention requiring a gemini amine surfactant to act as a structure directing agent when reacting with orthosilicate as an essential feature. The surfactant can then be removed leaving the structural framework and the resulting composition can then be reacted with functional groups. There is no surfactant used or necessary in Poncelet ('646) and thus again there is no motivation for the combination. The skilled person would know that in Pinnavaia (165) the non-hydrolyzable constituent(s) borne by the silane coupling agent would interact with the surfactant and help to grow the expected aluminosilicate structure and that the number of non-hydrolyzable groups would not be important. In the present invention there is no expectation that the non-hydrolyzable groups of the silane coupling agent would help to grow the isotropic amorphous structure.

The Examiner has referred newly to US 5,898,089 and in particular to col. 41 lines 33-55 as showing silane bearing precursors having one non-hydrolyzable group. In fact line 33 discloses tetraethylorthosilicate which has four hydrolyzable groups only and lines 34 and 35 disclose trimethylchlorosilane and chloromethyldimethyl chlorosilane, each with three non-hydrolyzable groups, although later examples do have only one non-hydrolyzable group. However there is no suggestion that a compound with only one non-hydrolyzable group is preferred – in fact, col. 4 lines 54 and 55 clearly state that the preferred silicon-containing compounds are tetraethyl orthosilicate (no non-hydrolyzable group) and poly(phenylmethyl) siloxane which is not a silicon alkoxide. It is not in dispute that silanes with only one non-hydrolyzable group are known, but there is no suggestion from either Pinnavaia (165) or Poncelet ('646) that there is any particular advantage in selecting a silane with only one nonhydrolyzable group from the various possibilities listed.

It is not clear if US 5,898,089 is intended to form part of the 35 USC 103 rejection. Clarification is respectfully requested.

It is gratefully noted that in paragraph 3, at page 9 of the application, Claim 24 was indicated as allowable. Nevertheless, for the reasons stated above it is respectfully urged that the other claims of this application are also allowable. It is urged that newly submitted Claim 25 is allowable for the same reason as Claim 24.

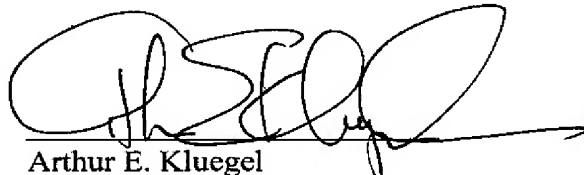


In paragraph 4 on page 10 of the Office Action, Claims 1-22 stand provisionally rejected on the ground of nonstatutory obviousness type double patenting. It is respectfully requested that the rejection be reconsidered and withdrawn in view of the Terminal Disclaimer that accompanies this amendment.

In view of the foregoing remarks and amendment, the claims 1-8, 10-22, 24, and 25 are now deemed allowable and favorable action is courteously solicited. Therefore, it is respectfully requested that the rejections under 35 USC 103, 35 USC 112 and obviousness type double patenting, be reconsidered and withdrawn and that an early Notice of Allowance be issued in this application.

Should the Examiner consider that additional amendments are necessary to place the application in condition for allowance, the favor is requested of a telephone call to the undersigned counsel for the purpose of discussing such amendments.

Respectfully submitted,

A handwritten signature in black ink, appearing to read 'Arthur E. Kluegel', with a long horizontal flourish extending to the right.

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**If the Examiner is unable to reach the Applicant(s) Attorney at the telephone number provided, the Examiner is requested to communicate with Eastman Kodak Company Patent Operations at (585) 477-4656.**